

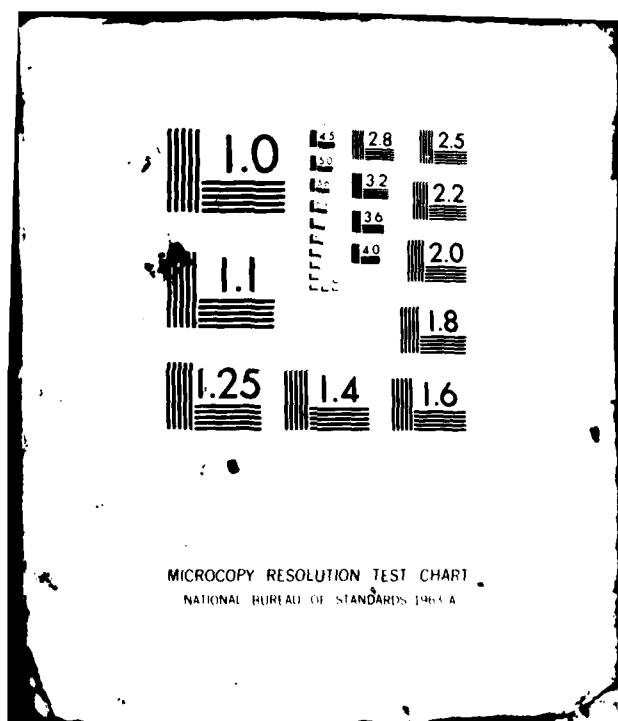
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PHYSICS OF ATOMIC FREQUENCY STANDARDS BASED ON STIMULATED RESONANCE RAMAN SCATTERING

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20. Abstract (Continued)

electronic state of sodium is discussed, and historical background on the Raman effect is presented. The lineshape and linewidth of the stimulated Raman transition are calculated by solving the Schrödinger equation for the atom interacting with two classical optical fields. The Raman line is shown to be transit-time broadened and to have no contribution from the broad resonant intermediate state. A version of the Ramsey separated-oscillatory-field method for reducing transit-time broadening, applicable to stimulated Raman scattering, is described and its effect on the linewidth is calculated. The influence of other possible broadening mechanisms, including Doppler broadening, laser jitter, and power broadening (optical Stark broadening) is determined, and it is shown that passive linewidths of kilohertz can be expected. Finally, the sources of Raman lineshifts contributing to frequency errors are mentioned.

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Preface

This work is an outgrowth of an ongoing research program at RADC on laser-assisted atomic frequency standards. The program is being carried out with the cooperation of the Massachusetts Institute of Technology's Research Laboratory for Electronics. We are greatly indebted to our colleagues Clare C. Leiby, Jr., of RADC and Professor Shaoul Ezekiel, Professor John E. Thomas, and Philip R. Hemmer of MIT for extensive discussions relating to the conceptual design of an atomic frequency standard based on resonant stimulated Raman scattering and to the ongoing experiments at RADC. We are grateful to our superiors at RADC, especially Dr. Andrew C. Yang, and to Dr. Thomas Walsh of AFOSR for encouragement and support. One of us (C.R.W.) gratefully acknowledges support from the University Resident Research Program (URRP) administered by AFOSR.



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Physics of Atomic Frequency Standards Based on Stimulated Resonance Raman Scattering

1. BACKGROUND

In this report, we analyze an attractive possibility for achieving high precision frequency standards based on stimulated resonance Raman scattering in atomic and molecular beams. We have proposed the use of this effect in frequency standards, as well as a first embodiment in a sodium atomic beam as an outgrowth of an ongoing RADC program.^{1, 2} The presentation will be tutorial in nature and will emphasize the physics of the interaction relevant to device operation, while avoiding many important engineering considerations.

The standards being considered rely neither on inhomogeneous magnets for polarizing or analyzing the atomic or molecular beam nor on hot-wire detectors for atomic detection. Rather they employ lasers for state preparation and detection of transitions, as well as for the basic interaction. Through a stimulated Raman process, the frequency difference of two lasers striking the atomic or molecular beam simultaneously in the interaction region is stabilized to a long-lived atomic or molecular transition. The difference of the two laser frequencies, and hence

(Received for publication 19 October 1981)

1. Ezekiel, S., Leiby, Jr., C.C., Picard, R.H., Willis, C.R., and Hackel, R.P. (1980) *Laser Stimulated Raman Molecular Beam Time and Frequency Standard*, allowed U.S. patent application No. 150,523, filed 16 May 1980.
2. Thomas, J.E., Ezekiel, S., Leiby, Jr., C.C., Picard, R.H., and Willis, C.R. (1981) *Opt. Lett.* 6:298.

the frequency output by the atomic/molecular portion of the standard, may vary from the microwave to the infrared region of the spectrum, depending on spacing of energy levels involved in the transition.

We concentrate in this report on factors affecting the precision of the frequency standards. For definiteness, we envision stimulated Raman scattering between the hyperfine levels ($F=1$ and $F=2$) in the ground electronic state ($3^2S_{1/2}$) of the sodium atom, separated by the microwave (L-band) frequency 1.772 GHz. For the moment, we ignore any subtleties introduced by the magnetic substates ($M_F = 0, \pm 1$ for the $F=1$ level and $M_F = 0, \pm 1, \pm 2$ for the $F=2$ level). This transition is the sodium transition analogous to the hyperfine transition ($F=4 \rightarrow F=3$) at 9.193 GHz induced in the $6^2S_{1/2}$ electronic ground state of cesium in the usual cesium atomic clock. To enhance the Raman cross section, we work near optical resonance of the two laser beams with the $3^2P_{1/2}$ or $3^2P_{3/2}$ excited state of sodium. The single-photon emission between the 3^2P levels and the ground state gives rise to the very intense characteristic yellow doublet D lines (D_1 and D_2) of sodium.

The history of the Raman effect began fifty years ago. The existence of scattered light with frequency shifted by an amount corresponding to the energy difference between two quantum states was discovered in 1928 by Raman and Krishnan³ in liquid solutions and by Landsberg and Mandelstam⁴ in solids. This discovery led to ordinary or spontaneous Raman scattering spectroscopy, which today is used mostly as a diagnostic tool for studying the vibrational energy levels of molecules and of optical-branch lattice vibrations in crystals and for identifying unknown samples or trace constituents. A spectral analysis of the scattered radiation reveals the existence of frequencies shifted down (Stokes scattering) or up (anti-Stokes scattering) by increments equal to vibrational frequencies of the material irradiated. A second weak beam of light applied to the sample at the Stokes-shifted (or anti-Stokes-shifted) frequency will experience gain. This is the phenomenon of stimulated Raman scattering first observed after the invention of the laser by Woodbury and others⁵ in 1962, in conjunction with vibrational transitions in organic laser Q-switching liquids. Of course, it is not necessary that the initial and final states of the scattering sample be different vibrational levels, as they were historically. Any two quantum states of the sample having the same parity will do.

In the quantum theory of radiation, Raman scattering appears as a two-photon process involving the $\mathbf{j} \cdot \mathbf{A}$ term of the interaction Hamiltonian, where \mathbf{j} is the

3. Raman, C. V., and Krishnan, K. S. (1928) Nature (London) 121:501.
4. Landsberg, G., and Mandelstam, L. (1929) Naturwissenschaften 16:557, 772.
5. Woodbury, E. J., and Ng, W. K. (1962) Proc. IRE 50:2367; Eckhardt, G., Hellwarth, R. W., McClung, F. J., Schwarz, S. E., Weiner, D., and Woodbury, E. J. (1962) Phys. Rev. Lett. 9:455.

polarization current density. Since this interaction is linear in the field vector potential \underline{A} , it causes transitions involving two photons through the agency of intermediate states of the scattering sample and the light field which differ from initial and final states by having only one photon absorbed or emitted. The Raman cross section will involve a sum over all possible intermediate states. Energy need not be conserved in the intermediate state, but only in the over-all transition from initial to final state. However, the proximity of the intermediate-state energy to an actual atomic resonance leads to a large enhancement of the Raman cross section similar to the enhancement of Rayleigh scattering near an atomic resonance (resonance fluorescence). This enhancement of the Raman effect is known as resonance Raman scattering. In the resonance Raman effect, there is only one intermediate state of any consequence — the resonant one — and the sum over intermediate states reduces to a single term. Hence, one need deal only with three energy eigenstates, the initial, the final, and the one intermediate state of consequence. It follows that the physics of resonance Raman scattering is inextricably tied up with the physics of material systems having three quantum states, or three-level systems.⁶ In Section 2, we will analyze stimulated resonance Raman scattering in terms of the semiclassical radiation theory of an optical field interacting with three-level systems, referred to hereafter as three-level "atoms."

Before proceeding, we should point out an important characteristic of resonance Raman scattering, namely, it cannot be described as absorption of the incident photon followed by emission of the scattered photon. This was first demonstrated convincingly, in conjunction with resonance fluorescence, about 1930 by Placzek⁷ and by Weisskopf.⁸ Rather, one must view the transfer of the atom from initial to final state, accompanied by destruction of an incident photon and creation of a scattered photon, as a single quantum process. In other words, transfer of atoms from the initial to the final state is not a two-step process, comprised of successive single-photon interactions, but a single-step coherent two-photon interaction. An important practical consequence of this is that the atom resides in the intermediate state for a negligible amount of time and, consequently, the linewidth of the upper level, which can be very large, does not adversely affect the width of the Raman resonance.⁹

In sodium, for example, the 3^2P intermediate state is separated by optical photon energies from the 3^2S initial and final states, and the $3P$ - $3S$ transition is

-
6. See, for example, Beterov, I. M., and Chebotaev, V. P. (1974) Progress in Quantum Electronics, Vol. 3, Part 1.
 7. Placzek, G. (1929) Z. Phys. 38:585.
 8. Weisskopf, V. (1931) Ann. Phys. (Leipzig) 9:23.
 9. Schlossberg, H. R., and Javan, A. (1966) Phys. Rev. Lett. 17:1242.

strongly allowed; consequently, the 3P state has a large natural linewidth of 10 MHz. On the other hand, the initial and final states are either the atomic ground state or levels lying close to the ground state whose transitions to the ground state are forbidden. Hence, there is the possibility of constructing atomic frequency standards based on optical photons having stabilities characteristic of microwave atomic standards. In Section 2, we show how the Raman process results in a narrow resonance whose width is independent of the intermediate state width.

2. THE STIMULATED RESONANCE RAMAN PROCESS

Consider the energy level diagram shown in Figure 1. An atom in level 1 moving at a speed v interacts simultaneously with two monochromatic optical fields, a pump field \underline{E}_p at frequency ω_p and a Stokes field \underline{E}_s at frequency ω_s . For simplicity, both fields are assumed to be zero outside the interaction region and perfectly uniform in the interaction region, which is of length d along the atomic velocity vector, such that the transit time through the region is $\tau = d/v$. The fields at the position of the atom are given by

$$\begin{aligned}\underline{E}_p(t) &= (1/2) \hat{e}_p E_{op} e^{-i\omega_p t + i\phi_p} + c. c. \\ \underline{E}_s(t) &= (1/2) \hat{e}_s E_{os} e^{-i\omega_s t + i\phi_s} + c. c. ,\end{aligned}\tag{1}$$

where \hat{e}_p and \hat{e}_s are unit polarization vectors for the waves and t is proportional to the distance the atom has penetrated into the interaction region. In order that there be no Doppler shift, the E field wave vectors both should be perpendicular to the atomic velocity.

The dynamics of the three-level atom is described by Schrödinger's equation with a phenomenological decay term added

$$i\hbar \frac{\partial \psi}{\partial t} = H\psi - \hbar \Gamma \psi ,\tag{2}$$

where H is the Hamiltonian for the atom and is the sum of an unperturbed part H^0 and a perturbation $V(t)$,

$$H = H^0 + V(t) ,\tag{3}$$

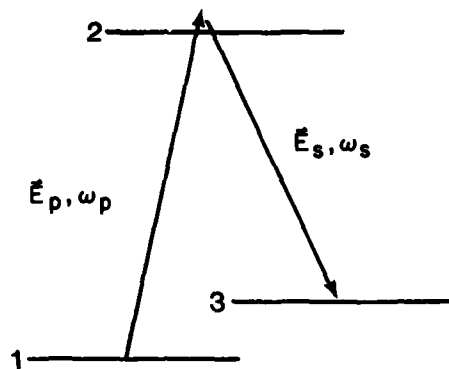


Figure 1. Energy Level Diagram Showing Stimulated Raman Interaction with Two Optical Fields

and Γ is a phenomenological relaxation operator describing atomic level decay. Since the atom is much smaller than an optical wavelength, the perturbation can be represented by a dipole interaction term,

$$V(t) = -\underline{\mu} \cdot [\underline{E}_p(t) + \underline{E}_s(t)] \quad (4)$$

where $\underline{\mu}$ is the atomic electric dipole moment. We now expand the wave function ψ in unperturbed atomic states $|m\rangle$ ($m = 1, 2, 3$), satisfying

$$H^0 |m\rangle = E_m |m\rangle; \quad (5)$$

that is,

$$\psi(t) = \sum_{m=1}^3 a_m(t) \exp(-i E_m t/\hbar) |m\rangle \quad (6)$$

We also assume that m th level amplitude decays at a rate $\gamma_m/2$, so that

$$\Gamma |m\rangle = (\gamma_m/2) |m\rangle \quad (7)$$

Substituting Eqs. (6) and (3) into Eq. (2) and using Eq. (5), Eq. (7), and the orthogonality of the eigenstates $|m\rangle$, $\langle m|n\rangle = \delta_{mn}$, we obtain the equation of motion for the probability amplitude in the interaction picture,

$$\dot{a}_n = -(i/\hbar) \sum_{m=1}^3 e^{i\omega_{nm}t} V_{nm}(t) a_m - (\gamma_n/2) a_n \quad (8)$$

In this equation

$$\omega_{nm} = (E_n - E_m)/\hbar \quad (9)$$

$$V_{nm}(t) = -\underline{\mu}_{nm} \cdot [E_p(t) + E_s(t)] \quad (10)$$

and $\underline{\mu}_{nm}$ is the transition moment (off-diagonal matrix element of the dipole operator); the superior dot denotes time differentiation. The square modulus of a_n gives the probability of being in the state $|n\rangle$, $W_n = |a_n|^2$, and the phase of a_n gives the phase shift for the state, whose time derivative is proportional to the energy shift of the level due to the interaction.

To obtain Eq. (8) in the explicit form to be used, we substitute from Eqs. (10) and (1) and neglect any nonresonant terms, including terms varying in time near twice the optical frequency such as $\exp[\pm i(\omega_p + \omega_{21})t]$ or $\exp[\pm i(\omega_s + \omega_{23})t]$. The result is

$$\dot{a}_1 + (\gamma_1/2)a_1 = (i/2\hbar) \mu_{21}^* E_{op}^* e^{-i\phi_p + i\Delta\omega_p t} a_2 \quad (11a)$$

$$\dot{a}_2 + (\gamma_2/2)a_2 = (i/2\hbar) \mu_{21} E_{op} e^{i\phi_p - i\Delta\omega_p t} a_1 + (i/2\hbar) \mu_{23} E_{os} e^{i\phi_s - i\Delta\omega_s t} a_2 \quad (11b)$$

$$\dot{a}_3 + (\gamma_3/2)a_3 = (i/2\hbar) \mu_{23}^* E_{os}^* e^{-i\phi_s - i\Delta\omega_s t} a_2 \quad (11c)$$

where

$$\Delta\omega_p \equiv \omega_p - \omega_{21} \quad (12a)$$

$$\Delta\omega_s \equiv \omega_s - \omega_{23} \quad (12b)$$

$$\mu_{21} = \hat{e}_p \cdot \underline{\mu}_{21}$$

$$\mu_{23} = \hat{e}_s \cdot \underline{\mu}_{23}$$

We assume initially at time $t = t_0$ when the atom enters the interaction region, there is no population in states $|2\rangle$ and $|3\rangle$,

$$a_m(t_0) = \delta_{m1} a_1(t_0) \quad . \quad (13)$$

and we solve the equations perturbatively for $a_3(t)$, to lowest possible order in the dipole moments μ_{mn} . The reader who is not interested in the details of the derivation may jump to Eq. (26).

In zero order only a_1 is nonzero and satisfies Eq. (11a) with $a_2 = 0$,

$$\dot{a}_1^{(0)} + (\gamma_1/2)a_1^{(0)} = 0 \quad , \quad (14)$$

whose solution is

$$a_1^{(0)}(t) = e^{-\gamma_1(t-t_0)/2} a_1(t_0) \quad (15)$$

Hence, $a_1^{(0)}(t)$ decays exponentially, resulting in a Lorentzian line broadening of level 1 with a width γ_1 . In a similar fashion, γ_2 and γ_3 will result in widths for levels 2 and 3, respectively. However, as discussed in Section 1, we will ignore the widths of levels 1 and 3, that is,

$$\gamma_1(t - t_0) \quad , \quad \gamma_3(t - t_0) \ll 1 \quad , \quad (16)$$

and assume that only level 2 has an appreciable width. Hence, Eq. (15) becomes simply

$$a_1^{(0)}(t) \cong a_1(t_0) \quad . \quad (17)$$

To first order in μ_{nm} , only a_1 is nonzero and satisfies Eq. (11b) with $a_1 = a_1^{(0)}$ and $a_3 = 0$,

$$\dot{a}_2^{(1)} + (\gamma_2/2)a_2^{(1)} = (i/2\hbar) \mu_{21} E_{op} e^{i\phi_p - i\Delta\omega_p t} a_1^{(0)}(t) \quad . \quad (18)$$

Solving Eq. (18) and using Eqs. (16) and (17), we obtain the probability amplitude for state 2,

$$a_2^{(1)}(t) = (i/2\hbar) \mu_{21} E_{op} e^{i\phi_p} a_1(t_0) [(\gamma_2/2)^{-1} - i\Delta\omega_p]^{-1} \\ \times \left[e^{-i\Delta\omega_p t} - e^{-\gamma_2(t-t_0)/2 - i\Delta\omega_p t_0} \right] . \quad (19)$$

If we look for time differences $t - t_0$ greater than the reciprocal linewidth γ_2^{-1} and assume that the frequency ω_p is within a linewidth γ_2 of ω_{21} —that is, that the Raman process is resonant,

$$\gamma_2(t - t_0) , \gamma_2/\Delta\omega_p \gg 1 \quad (20)$$

—the second term of Eq. (19) can be neglected, and the first simplifies to

$$a_2^{(1)}(t) \cong (i \mu_{21} E_{op}/\hbar \gamma_2) a_1(t_0) e^{i(\phi_p - \Delta\omega_p t)} . \quad (21)$$

By substituting Eq. (21) for a_2 into Eq. (11c) and using Eq. (16), we obtain an equation for a_3 , second-order in μ_{mn} ,

$$\dot{a}_3^{(2)} = (i/2\hbar) \mu_{23}^* E_{os}^* e^{-i\phi_s + i\Delta\omega_s t} a_2^{(1)}(t) ,$$

which can be integrated to yield

$$a_3^{(2)}(t) = \left[\Omega_{R,p} \Omega_{R,s}^* e^{i\chi} a_1(t_0)/2i\Delta_R \gamma_2 \right] \left[e^{-i\Delta_R t} - e^{-i\Delta_R t_0} \right] . \quad (22)$$

Here $\Omega_{R,p}$ and $\Omega_{R,s}$ are the Rabi frequencies corresponding to the pump and Stokes fields,

$$\Omega_{R,p} = \mu_{21} E_{op}/\hbar \quad (23)$$

$$\Omega_{R,s} = \mu_{23} E_{os}/\hbar .$$

χ is the phase difference between the two fields,

$$\chi = \phi_p - \phi_s . \quad (24)$$

and

$$\Delta_R \equiv \Delta\omega_p - \Delta\omega_s = \omega_p - \omega_s - \omega_{31} \quad (25)$$

Defining

$$g(x) \equiv e^{-ix} \sin x/x \equiv e^{-ix} \text{sinc } x \quad (26)$$

we can rewrite Eq. (22) in the form¹⁰

$$a_3^{(2)}(t) = -[\Omega_{R,p} \Omega_{R,s}^*(t - t_0)/2\gamma_2] a_1(t_0) e^{i(\chi - \Delta_R t_0)} g[\Delta_R(t - t_0)/2] \quad (27)$$

The transition probability to level 3, in second-order perturbation theory, is then

$$W_3^{(2)}(\tau) = |a_3^{(2)}(\tau)|^2 = |\Omega_{R,p} \Omega_{R,s} \tau/2\gamma_2 g(\Delta_R \tau/2)|^2 \quad (28)$$

where we have assumed the atom enters the field at $t_0 = 0$ and remains for a time τ . From Eqs. (16) and (20), we can summarize the conditions for the validity of Eq. (28) as

$$\gamma_1 \tau, \gamma_3 \tau \ll 1 \ll \gamma_2 \tau, \gamma_2/\Delta\omega_p \quad (29)$$

The properties of the stimulated Raman effect, in particular, the stimulated Raman lineshape, can be inferred from Eq. (28). It is clear from Eq. (26) that $W_3^{(2)}$ peaks at $\Delta_R = 0$. This is the Raman resonance condition, which states explicitly

$$\omega_p - \omega_s = \omega_{31} \quad (30)$$

or the difference between the pump and Stokes frequencies equals the atomic transition frequency between initial and final state. Moreover, as the optical frequency difference is detuned, the atomic response falls off as the square of the sinc function. Since $\text{sinc}^2 1.39 = 1/2$, it follows from Eq. (28) that the Raman linewidth (FWHM) is

$$\Gamma_R = 2(1.39)/\pi\tau = 0.885/\tau \quad (31)$$

10. The reader who has not followed the derivation above should refer to Eqs. (23) to (26) for the definitions of symbols occurring in this equation.

One concludes that the Raman line is transit-time broadened and insensitive to the broad linewidth of level 2 (10 MHz in the case of sodium). In a very real sense, the atom responds as if it had interacted with a beam of microwaves of frequency $\omega_\mu = \omega_p - \omega_s$. A beam of sodium atoms emerging from an oven at a temperature of 673K (400C) has most probable speed $v = 9 \times 10^4$ cm/sec. When the atoms traverse an interaction region of length $d = 1$ mm, Eq. (31) predicts a Raman linewidth of $\Gamma_R = 800$ kHz, much less than the 10 MHz value of γ_2 .

By increasing the length of the interaction region, the Raman width decreases in inverse proportion. Nevertheless, there is a limit to the amount of narrowing to be achieved in this way, if for no other reasons than that a finite amount of laser power is available. In Section 3 we examine another method of decreasing the transit-time linewidth of the Raman resonance.

3. THE RAMSEY METHOD IN STIMULATED RAMAN EMISSION

The experiment to detect Raman scattering described in Section 2, whose lineshape and width are given in Eqs. (28) and (31), is analogous to Rabi's resonance method in magnetic resonance.^{11, 12} In 1949 Ramsey^{13, 12} proposed a method of beating transit time broadening in molecular beam magnetic resonance experiments, based on the use of separated oscillating fields or separated interaction regions. The Ramsey method has also been applied successfully in optical resonance in the areas of saturated absorption spectroscopy and Doppler-free two-photon absorption spectroscopy, starting with the work of Baklanov and others^{14, 15} in 1976.

To apply the Ramsey method in stimulated Raman scattering spectroscopy,² we let the \underline{E}_p and \underline{E}_s together interact with the atomic beam in two regions of length d separated by a much larger distance $L-d$ (Figure 2). The distance L corresponds to a transit time $T = L/v$. The atomic transition to level 3, described by Eq. (27), can occur in either region A or region B, the upstream and

11. Rabi, I.I., Zacharias, J.R., Millman, S., and Kusch, P. (1938) Phys. Rev. 53:318; Kellogg, J.M.B., Rabi, I.I., Ramsey, N.F., and Zacharias, J.R. (1939) Phys. Rev. 55:728.
12. See also Ramsey, N.F. (1956) Molecular Beams (Clarendon Press, Oxford).
13. Ramsey, N.F. (1949) Phys. Rev. 76:996.
14. Baklanov, Ye.V., Dubetsky, B.Ya., and Chebotaev, V.P. (1976) Appl. Phys. 9:171; 11:201.
15. Bergquist, J.C., Lee, S.H., and Hall, J.L. (1977) Phys. Rev. Lett. 38:159.

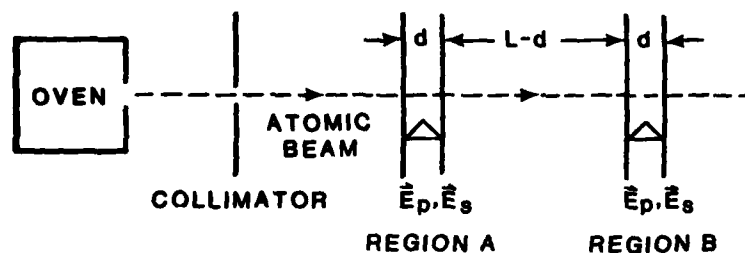


Figure 2. Experimental Setup for Applying Ramsey Method to Stimulated Raman Scattering by an Atomic Beam

downstream regions respectively.¹⁶ In perturbation theory the transition is weak in each region, and the transition amplitude small; if we observe the atom after it has traversed both regions, the total transition amplitude is the coherent sum of the individual transition amplitudes for each region.

Denote the amplitudes for undergoing transitions in regions A and B by subscripts on a_3 . Let $t = t_0 = 0$ when the atom enters region A and $a_1(0) = 1$. Equation (27) then yields

$$a_{3,A}^{(2)}(\tau) = -(\Omega_{R,p} \Omega_{R,s}^* \tau / 2\gamma_2) e^{i\chi_A} g(\Delta_R \tau / 2) \quad (32)$$

where the subscript A has been added to χ to allow the phase difference of the two optical waves to be different in the two regions. This amplitude then propagates freely until time $t = T + \tau$ when it is coherently combined with $a_{3,B}^{(2)}(T + \tau)$. We assume stronger, yet still realistic, conditions on γ_1 and γ_3 than in Eq. (29),

$$\gamma_1 T, \gamma_3 T \ll 1 \quad (33)$$

so that a_3 and a_1 do not change when they propagate freely. Thus we have

$$a_{3,A}^{(2)}(T + \tau) = a_{3,A}^{(2)}(\tau) \quad (34)$$

Similarly, to find the amplitude for undergoing a transition in region B, we let $t_0 = T$ and $a_{1,B}(T) = a_1(0) = 1$, obtaining

16. The A and B regions defined here have no relation to the A (state preparation) and B (detection) regions of a standard atomic clock. In fact, the regions called A and B here, taken together, correspond to the C, or interaction, region of an atomic clock.

$$a_{3,B}^{(2)}(T + \tau) = -(\Omega_{R,p} \Omega_{R,s}^* \tau / 2\gamma_2) e^{i(\chi_B - \Delta_R T)} g(\Delta_R \tau / 2) . \quad (35)$$

The transition probability to level 3 is given by

$$W_3^{(2)}(T + \tau) = |a_{3,A}^{(2)}(T + \tau) + a_{3,B}^{(2)}(T + \tau)|^2 ; \quad (36)$$

since the phase factors in Eqs. (32) and (35) differ in a frequency-dependent manner, we expect the lineshape to differ from the $\text{sinc}^2 x$ lineshape for the Rabi method in Eq. (28). Substituting from Eqs. (32), (34), and (35) into Eq. (36), we note that the Raman lineshape in the Ramsey method is given by

$$W_3^{(2)}(T + \tau) = |(\Omega_{R,p} \Omega_{R,s} \tau / \gamma_2) g(\Delta_R \tau / 2)|^2 \{ [1 + \cos(\Delta_R T + \chi_{AB})] / 2 \} . \quad (37)$$

where $\chi_{AB} = \chi_A - \chi_B$. From Eqs. (31) and (29) we may recapitulate the conditions under which this expression is valid,

$$\gamma_1 T, \gamma_3 T \ll 1 \ll \gamma_2 \tau, \gamma_2 / \Delta\omega_p . \quad (38)$$

The Ramsey lineshape is identical to the Rabi lineshape except for the factor in curly brackets. As the field frequency is tuned, this factor varies between 0 and 1 at a rate which is rapid compared to the factor $|g|^2$ because $T \gg \tau$. Hence, the Rabi lineshape is modulated by cosinusoidal Ramsey fringes due to the interference between the transition amplitudes in the two regions. Looked at in another way, the first (constant) term in Eq. (37) represents the sum of the transition probabilities in regions A and B, which are equal in this approximation, while the second (sinusoidally varying) term is a cross term in the individual amplitudes.

If we carefully control the low-frequency phase difference between \underline{E}_p and \underline{E}_s and arrange it to be the same in regions A and B, then $\chi_{AB} = 0$. In this case the central fringe peaks at $\Delta_R = 0$, and the width (FWHM) of the central fringe is

$$\Gamma_R' = (2T)^{-1} . \quad (39)$$

For the typical sodium atom of Section 2 and $L = 20$ cm, this results in $\Gamma_R' = 2$ kHz. By placing the interaction regions farther apart, we reduce the Raman linewidth in the Ramsey method yet further. Ultimately, our ability to reduce the linewidth

in this way is limited only by practical constraints on the length of the apparatus and volume of the vacuum system; by the mean-free path of the atoms in the beam region; and by the drop in atomic beam flux density at region B for large separations, combined with the finite amount of available laser power.

Comparing Eqs. (37) and (31) we see that the reduction in the linewidth by going to the Ramsey method is $\Gamma_R'/\Gamma_R \sim \tau/T = d/L$. For the typical values of d and L assumed above, we find $\Gamma_R'/\Gamma_R = 1/200$. Since the interacting atoms are in the form of an atomic beam, the transition probability given by Eq. (37) or (28) must be averaged over the speed distribution in the atomic beam. However, this averaging will not alter significantly the values Γ_R' and Γ_R of the linewidth, and we do not consider its effect on transit-time broadening.

In this section and Section 2 we have examined the influence of one broadening mechanism — transit-time broadening, a result of the short time which the atoms spend in the laser fields — on the Raman process. We have examined transit-time broadening in the Rabi method and in the Ramsey separated-field method where its effect is diminished. In the next section we examine other sources of broadening.

4. BROADENING MECHANISMS IN STIMULATED RAMAN EMISSION

Among the other possible broadening mechanisms that could affect the stability of atomic frequency standards based on stimulated Raman emission, in addition to transit-time broadening, are Doppler broadening, atomic level broadening, laser jitter, and power broadening (or optical Stark broadening). Line broadening enters into the frequency stability since the Allan variance $\sigma^2(2, \tau, \tau)$, the usual measure of fractional frequency stability, is given by

$$\sigma = (\Gamma_T/\nu_0)/(S/N) \quad (40)$$

where S/N is the signal-to-noise ratio, ν_0 the transition frequency, and Γ_T the total linewidth. Hence, larger linewidths require larger signal-to-noise ratios to obtain a given fractional frequency stability. We consider four sources of broadening as mentioned in the foregoing discussion.

4.1 Doppler Broadening

When a Raman transition with a moving atom occurs, the resonance frequency of the effective microwave field which the atom sees is shifted from

$$\omega_\mu = \omega_p - \omega_s$$

to

$$\omega_{\mu} = \omega_p - \omega_s - \vec{k}_{\mu} \cdot \vec{v} \quad (41)$$

where \vec{k}_{μ} is the effective microwave wave vector

$$\vec{k}_{\mu} = \vec{k}_p - \vec{k}_s$$

This is the first-order Doppler effect. By keeping \vec{k}_{μ} orthogonal to \vec{v} we can make the Doppler frequency shift equal to zero. However, with an atomic beam it is impossible to do this for all atoms, since the beam has a nonzero divergence. There is a residual shift due to the radial component of the beam velocity, which, for small beam divergence θ_0 , takes on a maximum value

$$v_r = u \theta_0$$

When the velocity average is performed, the net effect of all the shifts is to broaden the Raman transition, leading to a Doppler linewidth

$$\Gamma_D = [(\ln 2)^{1/2} / \pi] k_{\mu} u \theta_0 \quad (42)$$

For the 1.77 GHz hyperfine transition of sodium and an atomic beam divergence of 1 mrad, one easily finds $\Gamma_D = 5$ Hz. Hence, this source of broadening is quite negligible compared to transit-time broadening.

4.2 Atomic Level Broadening

A second potential source of broadening is the linewidths of the initial and final states 1 and 3 of the atom. These were denoted γ_1 and γ_3 in the Eqs. (11) and quite properly neglected in Eq. (16). The radiative lifetimes of levels 1 and 3 are extremely long, and there is no collision broadening of the levels in an atomic beam.

4.3 Laser Jitter

The Raman resonance can be broadened not only by the widths of atomic states 1 and 3, but also by the spectral width of the light incident on the atom. If we remove the requirement that the laser light fields be monochromatic, then Eqs. (1) are replaced by

$$\underline{E}_p(t) = (1/2) \hat{e}_p E_{op} e^{-i\omega_p t - i \int_0^t \delta\omega_p(t') dt' + i\phi_p} + c. c. \quad (43)$$

$$\underline{E}_s(t) = (1/2) \hat{e}_s E_{os} e^{-i\omega_s t - i \int_0^t \delta\omega_s(t') dt' + i\phi_s} + c. c.$$

Here the instantaneous frequencies $\delta\omega_p$ and $\delta\omega_s$ are random functions of time accounting for frequency jitter of the two light beams. The difference of the phase of E_p and E_s in Eq. (43) enters the calculation of the Raman transition amplitude $a_3^{(2)}$. Since $\delta\omega_p$ and $\delta\omega_s$ are statistically independent processes, the variance of the difference frequency is the sum of the variances of $\delta\omega_p$ and $\delta\omega_s$ and the rms contribution to the linewidth is

$$\Gamma_J = [\langle \delta\omega_p^2 \rangle + \langle \delta\omega_s^2 \rangle]^{1/2} \quad (44)$$

where the angular brackets denote statistical averaging. Since $\langle \delta\omega_p^2 \rangle^{1/2}$ can be hundreds of kilohertz, laser jitter would result in an unacceptably large linewidth completely dominating the transit-time width.

Nevertheless, there is a rather simple way to eliminate the effect of laser jitter, and that is to arrange for the jitter in the two laser fields to be correlated. This is accomplished by locking the two fields to a common reference cavity or, more simply, if the frequency difference is in an accessible region such as the microwave region, by deriving ω_p from the laser frequency ω_s through electro-optic or acousto-optic shifting.¹⁷ The acousto-optic shifting in a Bragg diffraction cell is particularly efficient and is convenient to carry out at the sodium transition frequency of 1.77 GHz. Letting $\delta\omega_l(t)$ be the instantaneous laser frequency due to jitter, we find

$$\omega_s(t) = \omega_l + \delta\omega_l(t)$$

$$\omega_p(t) = \omega_l + \omega_\mu + \delta\omega_l(t) \quad .$$

17. Leiby, Jr., C. C., and Ezekiel, S., Laser Excited Molecular Beam Time and Frequency Standard, allowed U.S. patent application No. 134,358, filed 27 March 1980.

When the difference frequency $\omega_s - \omega_p$ is formed, the laser jitter drops out of the problem,

$$\omega_s(t) - \omega_p(t) = \omega_\mu \quad ,$$

independent of $\delta\omega_l(t)$.

To determine the effect of laser jitter in a more precise manner, we can re-derive equations of motion for the probability amplitudes $a_n(t)$ as we did in Section 2, but using Eqs. (43) rather than Eqs. (1) for the optical fields. The result is Eqs. (11) with ϕ_i replaced by $\phi_i + \delta\phi_i(t)$ ($i = p, s$), where we have defined

$$\delta\phi_i(t) = - \int_0^t \delta\omega_l(t') dt' \quad . \quad (45)$$

If we carry out a derivation analogous to that of Eq. (27), we find that Eq. (19) is replaced by

$$a_2^{(1)}(t) = (i/2)\Omega_{R,p} e^{i\phi_p} a_1(t_0) \int_{t_0}^t \exp[-\gamma_2(t-t')/2 - i\Delta\omega_p t' + i\delta\phi_p(t')] dt' \quad . \quad (46)$$

while Eq. (22) is replaced by

$$a_3^{(2)}(t) = -(1/4)\Omega_{R,p} \Omega_{R,s}^* a_1(t_0) e^{i\chi} \int_{t_0}^t dt' e^{-i\Delta_R t'} \\ \times \int_{t_0}^{t'} dt'' \exp\{-\gamma_2(t'-t'')/2 + i[\delta\phi_p(t'') - \delta\phi_s(t')] \} \quad . \quad (47)$$

In deriving Eq. (47) we have used the approximations in Eqs. (16) and (20).

Assume now that the phases of the pump and Stokes waves are correlated, so that

$$\delta\phi_s(t) = \delta\phi_p(t) \equiv \delta\phi(t)$$

and

$$\delta\omega_s(t) = \delta\omega_p(t) \equiv \delta\omega_l(t) .$$

Then, for very slowly varying phases, we can expand $\delta\phi$ in a Taylor series,

$$\delta\phi(t'') - \delta\phi(t') \cong \dot{\delta\phi}(t')(t'' - t') = -\delta\omega_l(t')(t'' - t') , \quad (49)$$

and integrate Eq. (47) to obtain

$$a_3^{(2)}(t) = -(\Omega_{R,p} \Omega_{R,s}^* a_1(t_0) e^{ix/2\gamma_2}) \int_{t_0}^t dt' e^{-i\Delta_R t'} [1 - 2i\delta\omega_l(t')/\gamma_2]^{-1} . \quad (50)$$

It follows from Eq. (50) that if $\delta\omega_l \ll \gamma_2$, laser jitter has a negligible effect on the Raman transition.

The final integration in Eq. (50) cannot be carried out. Moreover, the series expansion of Eq. (49) is valid only in the limit $\gamma_2 \tau_c \gg 1$, where τ_c is the correlation time of the frequency fluctuations. An approach which is not restricted to this limit consists in averaging the random process $\delta\omega_l(t)$ over an appropriate probability density functional. To be correct, we should average the transition probability¹⁸ $W_3^{(2)} = |a_3^{(2)}|^2$; however, we obtain qualitatively correct results by averaging the transition amplitude in Eq. (47),

$$a_3^{(2)} = -(1/4) \Omega_{R,p} \Omega_{R,s}^* a_1(t_0) e^{ix} \int_{t_0}^t dt' e^{-i\Delta_R t'} \int_{t_0}^{t'} dt'' e^{-\gamma_2(t'-t'')/2} Q[l;t'',t'] , \quad (51)$$

where

$$Q[\lambda(t);t'',t'] \equiv \left\langle \exp \left[i \int_{t''}^{t'} dt \lambda(t) \delta\omega_l(t) \right] \right\rangle \quad (52)$$

18. Picard, R. H., and Willis, C. R. (to be published).

is the characteristic or moment-generating functional for the random process $\delta\omega_l(t)$.

For typical sources of laser jitter, such as vibration of a laser-cavity mirror, it is reasonable to assume that $\delta\omega_l(t)$ is a stationary, zero-mean, Gaussian process; then Q is determined solely by its second moment or correlation function

$$C(t_1 - t_2) \equiv \langle \delta\omega_l(t_1) \delta\omega_l(t_2) \rangle \quad (53)$$

through the relation¹⁹

$$Q[\lambda(t); t'', t'] = \exp \left\{ -(1/2) \int_{t''}^{t'} dt_1 \int_{t''}^{t'} dt_2 C(t_1 - t_2) \lambda(t_1) \lambda(t_2) \right\}. \quad (54)$$

After setting $\lambda(t) = 1$, using stationarity in the form $C(-\tau) = C(\tau)$, and changing the order of integration, one notes that Eq. (54) yields

$$Q[1; t'', t'] = \exp \left\{ - \int_0^{t' - t''} d\tau (t' - t'' - \tau) C(\tau) \right\}. \quad (55)$$

Notice that, with $\lambda(t) = 1$, Q is dependent only on the difference of its time arguments $t' - t''$, so that we may represent it by $Q[1; t' - t'']$.

Equations (51) and (55) comprise the complete solution of our problem. Once the correlation function $C(\tau)$ is specified, we can compute the characteristic functional Q and use it to determine the transition amplitude $a_3^{(2)}$. To determine the effect of jitter on the width of the atomic response, we can change variables of integration in Eq. (51) to $\tau' \equiv t'' - t_0$ and $\tau'' \equiv t' - t''$, then interchange the order of integration, with the result

$$a_3^{(2)} = -(1/4) \int_0^{t-t_0} d\tau'' e^{-[(\gamma_2/2) + i\Delta_R] \tau''} Q[1; \tau''] \int_0^{t-t_0-\tau''} d\tau' e^{-i\Delta_R \tau'}. \quad (56)$$

Since τ'' is limited to $\lesssim \gamma_2^{-1}$, it can be ignored compared to $t - t_0$, by Eq. (20), and the inner integral has the value $(t - t_0)g[\Delta_R(t - t_0)/2]$. If we restrict ourselves

19. Middleton, D. (1960) Statistical Communication Theory, McGraw-Hill, New York, pp. 372-373.

to detunings satisfying $\Delta_R \ll \gamma_2$, a very weak restriction, we may also ignore Δ_R in the exponent of the τ'' integral, so that finally $a_3^{(2)}$ is given by Eq. (27) with the right member multiplied by the factor

$$(\gamma_2/2) \int_0^{t-t_0} d\tau'' e^{-\gamma_2 \tau''/2} Q[1; \tau''] \quad (57)$$

Further details on the influence of laser jitter, including the important limiting cases of slow modulation and fast modulation, will be presented elsewhere.¹⁸ However, important qualitative conclusions on laser jitter of arbitrary strength and time scale can be drawn from Eq. (57). Since the factor in Eq. (57) is generally smaller than unity, jitter will lead to a small reduction of the transition amplitude. However, because Eq. (57) is real and independent of Δ_R , jitter will not affect the atomic lineshape or linewidth at all. Hence, laser frequency fluctuations, like the broad width of level 2, do not affect the Raman linewidth, as long as we correlate the jitter at the two frequencies in some fashion.

4.4 Power Broadening

Finally, we mention the influence of power broadening. When the field E_p is applied to the atom near resonance between levels 1 and 2, both levels split by an amount equal to the Rabi frequency $\Omega_{R,p}$ (optical Stark effect). If the splitting is small compared to the level widths, it is not resolved and appears as a broadening of the level by an amount $\Omega_{R,p}^2/2\pi\gamma_2$, when $\gamma_1, \Delta\omega_p \ll \gamma_2$. This process is illustrated in Figure 3(a). The interpretation of this broadening is that level 1 has acquired a width due to the coherent superposition of state 1 and the short-lived state 2 induced by the optical field. In a similar fashion, the Stokes field broadens levels 3 and 2, with the result that the Raman transition from 1 to 3 in the Rabi method will have a width due to power broadening [Figure 3(b)] given by²⁰

$$\Gamma_p = (\Omega_{R,p}^2 + \Omega_{R,s}^2)/2\pi\gamma_2 \quad (58)$$

Here it has been assumed that

$$\gamma_1, \gamma_3, \Delta\omega_p, \Delta\omega_s \ll \gamma_2 \quad (59)$$

For dipole moments typical of sodium, μ_{12} and $\mu_{32} \cong 6$ Debye (1 Debye = 10^{-18} esu), and light intensities $I_p = I_s = 50 \mu W/cm^2$, we find $\Gamma_p = 250$ kHz. This is

20. See Bloembergen, N., and Levenson, M.D. (1976) in High-Resolution Laser Spectroscopy, K. Shimoda, Ed., Springer-Verlag, Berlin, p. 315.

comparable to the transit-time broadening Γ_R and shows that one must limit oneself to weak laser fields to avoid making power broadening the dominant broadening mechanism.

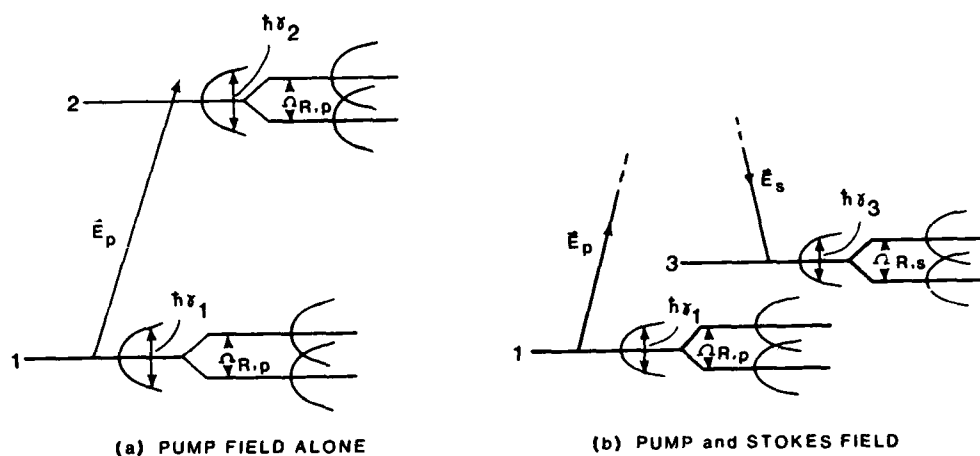


Figure 3. Effects of Optical Stark Splitting on Atomic Level Widths: (a) with pump field alone present; (b) with both pump and Stokes fields present

In the Ramsey method there is a dramatic reduction in power broadening from that calculated above for the Rabi method. To see this effect qualitatively, we recall from Eq. (37) that the Ramsey-method lineshape is a product of cosinusoidal interference fringes and an envelope given by the Rabi lineshape. The envelope is power broadened as in the previous paragraph. The fringes, which arise due to the transit of the atoms through the dark region between A and B, are not broadened directly. However, when the indirect effect of the envelope's broadening on the width of the central fringe is calculated,¹⁸ it is found that the latter is broadened by a factor of order $(d/L)^2$ times the value in Eq. (58). For the parameters assumed in Section 3, this reduction factor is of order 2×10^{-5} , and the resulting width is of order 10 Hz.

In passing, we note that, not only the linewidth, but also the signal-to-noise ratio, is laser power dependent. The latter goes up with the square root of the laser power, if the detection process is photon shot-noise-limited. However, this increase is not sufficient to overcome the increase in linewidth. The over-all result is that the Allan variance goes up with power, as is obvious from Eq. (40).

4.5 Implications for Frequency Standards

It is therefore to be concluded that, with proper precautions, none of the aforementioned broadening mechanisms compete seriously with transit-time broadening as an important constituent of the Raman linewidth in the Ramsey method. In particular, we have shown how the effect of laser jitter may be completely eliminated by correlating the frequency jitters of the pump and Stokes beams.

5. LINESHIFTS IN STIMULATED RAMAN EMISSION

In addition to the broadening introduced by the various physical phenomena discussed in Section 4, most of these phenomena also introduce a shift in the position of the Raman line. Determination of these shifts is especially important, since they introduce systematic errors into the frequency standard. Unlike line broadening, no amount of increase in signal-to-noise ratio will decrease the effect of the lineshift on the accuracy of the standard.

The main sources of Raman lineshifts are first-order and second-order (transverse) Doppler effect, differences in microwave phase $\phi_{\mu} \equiv \phi_p - \phi_s$ between the two interaction regions in the Ramsey method, and power-dependent atomic level shifts (light shifts). The subject of lineshifts is important enough to merit attention on its own. A separate paper will be published.¹⁸

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